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⑯ References cited :
EP-A- 0 384 944
EP-A- 0 405 088
PATENT ABSTRACTS OF JAPAN vol. 13, no.
571 (E-862)(3919) December 18, 1989 & JP-A-01
239 773
PATENT ABSTRACTS OF JAPAN vol. 12, no.
273 (E-639)(3120) July 29, 1988 & JP-A-63 053
858
PATENT ABSTRACTS OF JAPAN vol. 11, no.
306 (E-546)(2753) October 6, 1987 & JP-A-62
098 567
PATENT ABSTRACTS OF JAPAN vol. 13, no.
571 (E-862)(3919) December 18, 1989 & JP-A-01
239 774

⑯ References cited :

PATENT ABSTRACTS OF JAPAN vol. 13, no.
312 (E-788)(3660) July 17, 1989 & JP-A-01 084
578
PATENT ABSTRACTS OF JAPAN vol. 13, no.
46 (E-711)(3394) February 2, 1989 & JP-A-63
241 871

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Description**BACKGROUND OF THE INVENTION****Field of the Invention**

This invention relates to internally manifolded and internally reformed fuel cell stacks, and in particular, a method and process for sealing fully internally manifolded cell stacks with wet seals between only the electrolyte and metallic separator plates to provide long term stability while providing an internal reforming chamber separated from the anode chamber.

Generally, fuel cell electrical output units are comprised of a stacked multiplicity of individual cells separated by inert or bi-polar electronically conductive ferrous metal separator plates. Individual cells are sandwiched together and secured into a single stacked unit to achieve desired fuel cell energy output. Each individual cell generally includes an anode and cathode electrode, a common electrolyte tile, and a fuel and oxidant gas source. Both fuel and oxidant gases are introduced through manifolds to their respective reactant chambers between the separator plate and the electrolyte tile. The area of contact between the electrolyte and other cell components to maintain separation of the fuel and oxidant gases and prevent and/or minimize gas leakage is known as the wet seal. A major factor attributing to premature fuel cell failure is corrosion and fatigue in the wet seal area. This failure is hastened by corrosive electrolyte contact at high temperatures and high thermal stresses resulting from large temperature variations during thermal cycling of the cell causing weakening of the structure through intracrystalline and transcrystalline cracking. Such failures permit undesired fuel and/or oxidant gas crossover and overboard gas leakage which interrupts the intended oxidation and reduction reactions thereby causing breakdown and eventual stoppage of cell current generation. Under fuel cell operating conditions, in the range of about 500° to 700° C., molten carbonate electrolytes are very corrosive to ferrous metals which, due to their strength, are required for fuel cell housings and separator plates. The high temperature operation of stacks of molten carbonate fuel cells increases both the corrosion and thermal stress problems in the wet seal area, especially when the thermal coefficients of expansion of adjacent materials are different.

This invention provides fully internal manifolding of the fuel and oxidant gases to the individual cells of an assembled stack in a manner utilizing electrolyte/metal wet seals which, due to the design of the cell components, provides long term endurance and stability of fuel cell operation. This invention also provides internal manifolding for a separated reforming chamber for internal cell reforming of hydrocarbon containing fuels without poisoning of the reforming

catalyst. The endothermic reaction of reforming methane to carbon oxide and hydrogen is advantageously carried out within the cell stack.

5 Description of the Prior Art

Commercially viable molten carbonate fuel cell stacks may contain up to about 600 individual cells each having a planar area in the order of eight square feet. In stacking such individual cells, separator plates separate the individual cells with fuel and oxidant each being introduced between a set of separator plates, the fuel being introduced between one face of a separator plate and the anode side of an electrolyte matrix and oxidant being introduced between the other face of the separator plate and the cathode side of a second electrolyte matrix.

The emphasis in fuel cell development has been in external manifolding of the fuel and oxidant gases by using channel manifolds physically separable from the fuel cell stack. However, the inlets and outlets of each cell must be open to the respective inlet and outlet manifolds which must be clamped onto the exterior of the cell stack. To prevent electrical shorting, insulation must be used between the metal manifolds and the cell stack. External manifolding has presented serious problems in maintaining adequate gas seals at the manifold/manifold gasket/cell stack interface while preventing carbonate pumping within the gasket along the potential gradient of the cell stack. Various combinations of insulating the metal manifold from the cell stack have been used, but with the difficulty of providing a sliding seal which is gas tight and electrically insulating while being carbonate impermeable under high temperature molten carbonate fuel cell operating conditions, no satisfactory solution has been found. The problem of manifolding and sealing becomes more severe when larger number of cells and larger planar areas are used in the cell stack. When greater numbers of cells are used, the electrical potential driving the carbonate in the seal area along the height of the stack increases, and when the planar area of the cell increases, the linear tolerances of each component and the side alignment of each component becomes extremely difficult to maintain in order to maintain the mating surface sealed between the manifold/manifold gasket/and cell stack.

Cell stacks containing 600 cells can be approximately 10 feet tall presenting serious problems of required stiffness of external manifolds and the application of a clamping force required to force the manifold onto the cell stack. Due to the thermal gradients between cell assembly and cell operating conditions, differential thermal expansions, and the necessary strength of materials used for the manifolds, close tolerances and very difficult engineering problems are presented.

Conventionally, stacks of individual molten car-

bonate fuel cells have been constructed with spacer strips around the periphery of a separator plate to form wet seals and to provide intake and exhaust manifolds. Various means of sealing in the environment of the high temperature fuel cell wet seal area are disclosed in U.S. Patent 4,579,788 teaching the wet seal strips are fabricated utilizing powder metallurgy techniques; U.S. Patent 3,723,186 teaching the electrolyte itself is comprised of inert materials in regions around its periphery to establish an inert peripheral seal between the electrolyte and frame or housing; U.S. Patent 4,160,067 teaching deposition of inert materials onto or impregnated into the fuel cell housing or separator in wet seal areas; U.S. Patent 3,867,206 teaching a wet seal between electrolyte-saturated matrix and electrolyte saturated peripheral edge of the electrodes; U.S. Patent 4,761,348 teaching peripheral rails of gas impermeable material to provide a gas sealing function to isolate the anode and cathode from the oxidant and fuel gases, respectively; U.S. Patent 4,329,403 teaching graded electrolyte composition for more gradual transition in the coefficient of thermal expansion in passing from the electrodes to the inner electrolyte region; and U.S. Patent 3,514,333 teaching housing of alkali metal carbonate electrolytes in high temperature fuel cells by use of a thin aluminum sealing gasket. None of the above patents deal with sealing around internal fuel and oxidant in fuel cell stacks.

Gas sealing of a phosphoric acid fuel cell, which operates at about 150° to 220° C., by filling the pores of a porous material periphery of the cell constituents with silicon carbide and/or silicon nitride is taught by U.S. Patent 4,781,727; and by impregnating interstitial spaces in substrate plate edge is taught by U.S. Patents 4,786,568 and 4,824,739. The solution of sealing and corrosion problems encountered in low temperature electrolytic cells, such as bonding granular inert material with polytetrafluoroethylene as taught by U.S. Patent 4,259,389 gaskets of polyethylene as taught by U.S. Patent 3,012,086; and "O" ring seals taught by U.S. Patent 3,589,941 for internal manifolding of fuel only are not suitable for high temperature molten carbonate fuel cells.

U.S. Patent 4,510,213 teaches transition frames surrounding the active portion of the cell units to provide fuel and oxidant manifolds to the gas compartments of the individual cells, the manifolds not passing through the separators nor the electrolyte tiles of the cells. The transition frames require complicated insulating between adjacent cells and are made up of several separate and complicated components. U.S. Patent 4,708,916 teaches internal manifolding of fuel and external manifolding of oxidant for molten carbonate fuel cells wherein sets of fuel manifolds pass through electrodes as well as electrolytes and separators in a central portion and at opposite ends of the individual cells to provide shortened fuel flow paths.

The end fuel manifolds are in a thickened edge wall area of the separator plate while the central fuel manifolds pass through a thickened central region and sealing tape impregnated with carbonate or separate cylindrical conduit inserts are provided extending through the cathode.

Internal manifolding has been attempted wherein multiple manifold holes along opposite edges of the cell have been used to provide either co- or counter-current flow of fuel and oxidant gases. These manifold holes for fuel have been located in a broadened peripheral wet seal area along opposing edges, but the manifolds have been complicated structures exterior to the electrolyte or pass through at least one of the electrodes. However, adjacent manifold holes are used for fuel and oxidant which provides short paths across a short wet seal area and leakage of the gases as well as the necessarily broadened peripheral seal area undesirably reducing the cell active area.

Likewise, prior attempts to provide internal manifolding have used multiple manifold holes along broadened peripheral wet seal areas on each of all four edges of the cell to provide cross flow, but again short paths between adjacent fuel and oxidant manifold similar complicated structures and holes caused leakage of the gases and further reduced the cell active area.

When using gasification products as fuel, it is desirable to reform the hydrocarbonaceous components to enhance the hydrogen content of the fuel by internal reforming within the fuel cell stack. However, conventional reforming catalysts are known to be poisoned by molten carbonates electrolytes due to active sites being covered by a film of carbonates. See "Development of Internal Reforming Catalysts for the Direct Fuel Cell", Michael Tarjanyi, Lawrence Paetsch, Randolph Bernard, Hosein Ghezel-Ayagh. 1985 Fuel Cell Seminar, Tucson, Ariz., May 19-22, 1985. pgs. 177-181. Additional known problems causing failure in long term endurance of molten carbonate fuel cells also include deformation of the porous anode structure, corrosion of anode side hardware such as current collector, separator plate, and the like, by the molten carbonates electrolyte and electrolyte loss thereby, gas cross-over through the porous anode, and electrolyte loss by anode and cathode dissolution. There have been many attempts to solve one or more of these problems to provide long term fuel cell stability and endurance.

Increasing the hydrogen content of the fuel feed stream to the anode compartment of a fuel cell is taught by several patents. U.S. Patent 3,266,938 teaches a plurality of high temperature fuel cells arranged in series such that the spent gases from the anode compartment of the first fuel cell in the series is catalytically reformed exterior to the cell by an endothermic reforming reaction to produce additional hydrogen and then passed to the anode compartment

of a second cell in the series; the spent gases of the anode compartment of the second fuel cell is passed to a catalytic exothermic shift reaction exterior to the cell for further production of hydrogen for passage to the anode compartment of a third fuel cell in the series. The reforming and shift reactions are performed exterior to the fuel cells to provide greater hydrogen content to the fuel feeds to the anode compartments of the fuel cells. U.S. Patent 4,522,894 teaches increasing the hydrogen content of a liquid hydrocarbon feed by catalytic oxidation and steam reforming wherein use of thermal energy from the oxidation is used for reforming external to the fuel cell, to produce high hydrogen content in the fuel feed stream to the anode compartment of the fuel cell. U.S. Patent 3,488,226 teaches low temperature, low pressure steam reforming of liquid hydrocarbons to enhance hydrogen in the fuel feed for the anode compartment of molten carbonate fuel cells wherein the reforming is performed exterior to the fuel cell and acts as a heat sink for fuel cell produced heat. In one embodiment, the reforming catalyst may be placed in the fuel cell anode chamber. In either arrangement, the waste heat from the fuel cell is used directly to sustain the endothermic reforming reaction for the generation of hydrogen. U.S. Patent 4,702,973 teaches a dual compartment anode structure for molten carbonate fuel cells wherein the molten carbonates electrolyte is isolated from contaminated fuel gases and reforming catalysts by a hydrogen ion porous and electrolyte non-porous metallic foil.

SUMMARY OF THE INVENTION

This invention provides fully internally manifolded fuel cell stacks with reforming of hydrocarbonaceous fuel within the fuel cell stack to provide thermal energy for the endothermic reforming reaction while providing desired cooling of the fuel cell stack due to heating by the exothermic electrochemical reaction of the fuel cell. This invention is especially suitable for use in high temperature molten carbonate fuel cell stacks and provides separation of the reforming catalyst from the electrolyte, which, in the case of molten alkali carbonates, readily poisons the catalyst. The fully internally manifolded fuel cells of this invention are suitable for any cell having planar components, especially other high temperature fuel cells such as solid oxide fuel cells. A generally rectangular fuel cell stack is comprised of a plurality of fuel cell units, each fuel cell unit comprising an anode and a cathode, an electrolyte in contact on one side with the anode and in contact on the opposite side with the cathode, and a separator plate separating cell units between the anode of one cell and cathode of the adjacent cell forming an anode chamber between one side of the separator plate and the anode and a cathode chamber between the opposite side of the separator plate

and the cathode. The fuel cell units are stacked and provided with end plates having the same internal configuration as the separator plates forming half cells at each end and clamped to provide rigid structure to the fuel cell stack. Within the cell stack and interspersed between groups of about 5 to about 10 cell units reforming chambers may be formed by slightly modified separator plates wherein hydrocarbonaceous fuel and steam is supplied through an internal reaction gas manifold, passed over reforming catalyst in the reforming chamber, and the hydrogen enriched product gas passed to the internal fuel manifold of the fuel cell stack. In the fuel cell stacks of this invention, the electrolytes and separator plates have the same configuration and extend to the edge of the fuel cell stack, while the electrodes and current collectors do not extend to the edge of the fuel cell stack. The separator plates have a flattened peripheral wet seal structure extending to contact the electrolytes on each face of the separator plates completely around their periphery forming a continuous peripheral separator plate/electrolyte wet seal under cell operating conditions.

The electrolytes and separator plates have a plurality of aligned perforations in desired locations, each separator plate perforation being surrounded by a flattened manifold wet seal structure extending to contact the electrolyte on each face of the separator plate forming a separator plate/electrolyte manifold wet seal under cell operating conditions surrounding each perforation to form a gas manifold through each perforation and extending through the cell stack. Conduits or holes through the extended manifold wet seal structure provide gas communication between fuel manifolds and the anode chambers on one face of the separator plates and conduits or holes through the extended manifold wet seal structure provide gas communication between the oxidant manifolds and the cathode chambers on the other face of the separator plates. This structure provides fully internal manifolding of fuel and oxidant gases to and from each of the unit fuel cells in the fuel cell stack. In a similar manner, hydrocarbonaceous reaction gas and steam may be supplied to reforming chambers interspersed throughout the fuel cell stack and hydrogen enriched reformed product gas passed from each of the reforming chambers to the fuel manifold.

The end plates are configured similarly to the separator plates on their inner sides and are provided with means for supply and exhaust from each of the sets of manifolds of the fuel cell stack. External means of supplying and exhausting fuel gas, oxidant gas, and hydrocarbonaceous reaction gas to the appropriate sets of manifolds at the end plate connections may be provided by any means known to the art. By "sets of manifolds" we mean a first set makes up one or more fuel inlets, a second set one or more spent fuel outlets, a third set one or more oxidant in-

lets, a fourth set one or more spent oxidant outlets, and a fifth set one or more reaction gas inlets. The perforations through the separator plates and electrolytes forming the manifolds may be round, square, rectangular, triangular, or any other desired shape and size. While each such perforation is referred to as a single perforation, it may comprise baffling to provide desired gas distribution. Any number of manifolds may be provided through the separator plates and electrolytes as necessary to provide desired gas flow quantities and patterns across the active areas of the cell. It is important in this invention to provide full wet seals directly between the separator plate and electrolyte around each manifold with the edge of adjacent manifolds being separated by at least about 0.25 inch. This invention also provides a continuous peripheral wet seal directly between the separator plate and electrolyte exterior to the regions of the internal manifolds.

In one preferred embodiment, the separator plates in accordance with this invention are thin pressed metal plates provided with corrugations in the fully active fuel cell area and pressed to form on one face the full peripheral and the manifold wet seal structures with an upstanding wet seal structure welded to the opposite face of the separator plate to provide full peripheral and the manifold wet seals between the separator plate and electrolyte on opposite faces of the separator plates. Any structure may be used to provide the extended wet seal areas for forming wet seals directly between the separator plate and electrolyte, such as bars, strips formed by powder metallurgy techniques, and the like.

In a preferred embodiment the conduits or holes through the extended manifold wet seal structure providing gas communication between the manifold and the anode and cathode chambers as well as the reforming chamber may be openings provided by appropriately corrugated metal or may be holes through sheet metal or bar structures.

This invention provides simple wet seals between flattened, thin sheet metal structures and electrolyte, thereby providing assured sealing of one gas conduit from the adjacent gas conduit. This provides effective means for providing fully internally manifolded gas feed and removal from high temperature and corrosive fuel cells, such as, molten carbonate fuel cell stacks. Use of the structure of this invention also provides effective and varied means for providing carbonate to multi-cell stacks.

The structure of this invention provides fully internally manifolded reforming chambers interspersed in the fuel cell stack to reform hydrocarbonaceous fuel gas providing enhanced hydrogen fuel to the anode compartments while advantageously utilizing electrochemical produced thermal energy to drive the reforming reaction while cooling the fuel cell stack.

This invention provides a mass producible con-

figuration of the fuel cell components, particularly the separator plate and its cost effective fabrication. Use of the molten carbonate fuel cell units of this invention provides ease of assembly of the fuel cell stack and modularization for varying sizes of fuel cell stacks.

This invention also provides a process for production of electricity using the fully internally manifolded fuel cell stack with internal reforming, particularly, molten alkali metal carbonates fuel cell stacks.

BRIEF DESCRIPTION OF THE DRAWING

Further features of the invention will be apparent from the following detailed description of the invention read in conjunction with the drawing, wherein:

- 5 Fig. 1 is a schematic exploded side sectional view of a single cell along a diagonal to illustrate the principles of this invention;
- 10 Fig. 2 is an exploded perspective view of a single cell unit of a fuel cell stack in accordance with one embodiment of this invention;
- 15 Fig. 3 is a sectional side view of a peripheral wet seal area of a fuel cell according to one embodiment of this invention;
- 20 Fig. 4 is a sectional side view of a single cell unit showing opening from a fuel manifold conduit to the anode compartment;
- 25 Fig. 5 is a sectional side view of the cell unit shown in Fig. 4 showing opening from an oxidant manifold conduit to the cathode compartment;
- 30 Fig. 6 is a front face view of another embodiment of a manifold plate for a fully manifolded fuel cell stack according to this invention;
- 35 Fig. 7 is an opposite face view of the manifold plate shown in Fig. 6;
- 40 Fig. 8 is an enlarged cross-sectional view along 8-8 shown in Fig. 6;
- 45 Fig. 9 is an enlarged cross-sectional view along 9-9 shown in Fig. 6;
- 50 Fig. 10 is an enlarged cross-sectional view along 10-10 shown in Fig. 6; and
- 55 Fig. 11 is a schematic exploded perspective view of a portion of a fuel cell stack with an internal reforming chamber according to one embodiment of this invention.

DESCRIPTION OF PREFERRED EMBODIMENTS

This invention is directed toward fully internally manifolded fuel cell stacks. In preferred embodiments the electrolyte tile is penetrated by the manifold conduits and in specified areas the electrolyte contacts the separator plate to provide an electrolyte/separator plate peripheral wet seal for containment of fluids within the cell stack and an electrolyte/separator plate manifold seal for isolation of reactant compartments and for direction of fluids into and out of reactant compartments within individual molten

carbonate fuel cells and reforming chambers interspersed in the fuel cell stack. This invention preferably utilizes thin sheet separator plates having pressed seal areas extending from one face of the separator plate and thin sheet forms extending from the opposite face of the separator plate to form seal areas. The thin sheet seal areas afford limited flexibility and resilience to provide tight sealing.

Referring to Figure 1, a schematic exploded sectional view along a corner to corner diagonal of a single cell of a fuel cell stack according to this invention is shown which provides flow of fuel and oxidant gases fully internal to the cell stack. According to this embodiment, manifold holes are provided in the corner areas of the electrolyte which extends to the edge of the cell together with the cell separator plates. By contact between the electrolyte and the separator plate on each face forming conventional wet seals on each face around the periphery of the electrolyte, the containment of the fluids is maintained. Through desired openings providing fluid communication between manifold holes and anode and cathode compartments, the desired gaseous flow may be obtained while providing sealing of the manifold holes with conventional electrolyte/separator plate wet seals.

The matching manifold holes in the separator plates and electrolyte tiles form manifold conduits which are continuous for the entire height of the fuel cell stack for gas supply and exhaust. This invention provides that a manifold conduit extending to all cells in a fuel cell stack is supplied from a single external opening, whereas prior externally manifolded fuel cell stacks have required external openings to and from each individual fuel cell. The gases are fed to the fuel cell stack through an end plate which acts as one half cell and are exhausted through a similar end plate which acts as another half cell.

The manner in which fluids are fed to and withdrawn from fuel cell stacks can take on a wide variety of variations, the important aspect with respect to the present invention being that gas sealing is accomplished by sealing between the electrolyte tile and the separator plate in the conventional wet seal manner both around the periphery of the separator plate and in the gas manifold area as desired for conducting the gas to desired locations within each individual cell and the reforming chambers interspersed in the fuel cell stack.

As shown in Fig. 1, electrolyte 20 and separator plate 40 extend to the outer edge of the cell and are sealed to each other around their periphery in wet seal areas 23. In Fig. 1, the individual molten carbonate fuel cell unit is shown with anode 26 spaced from one face of separator plate 40 to provide an anode chamber fed by fuel manifold hole 24 as indicated by arrow 38. On the other face of separator plate 40 cathode 27 is spaced from separator plate 40 to form a cathode chamber in communication with oxidant

manifold holes 25 as indicated by arrow 39. Electrolyte 20 and separator plate 40 extend to the outer edge of the cell forming peripheral wet seal areas 23 which provide peripheral wet seals between the electrolyte and separator plate for containment of fluid. Fuel manifold wet seal area 45 and oxidant wet seal area 46 provide manifold sealing by electrolyte/separator plate wet seals and provide desired guidance of fluid to anode and cathode chambers on opposite sides of separator plate 40. No additional gaskets are used for sealing and the cell unit can accommodate a wide variety of carbonate addition techniques, including use of carbonate tapes. When carbonate tapes are used, the carbonate tapes and electrolyte matrix extend to the cell edges and although the inter-cell spacing decreases in proportion to the thickness of the carbonate tapes when they melt, sealing and conformity of all cell components is maintained at all times. During cell heat-up prior to carbonate tape melting, sealing is maintained around each manifold hole 24 and 25 because the carbonate tapes and the electrolyte matrix, such as LiAlO₂, extend adjacent to the respective sealing surfaces and contain a rubbery binder. During binder burn-out, which occurs prior to carbonate melt, gas flows are maintained and sealing is obtained. When the binder is burned off and the cell temperature raised to the melting point of the carbonate, the melting carbonate is absorbed by the porous LiAlO₂ tape and the electrodes. The inter-cell spacing decreases as the carbonate tapes melt but at all stages from room temperature to operating temperatures of about 650° C. cell sealing is maintained. The limited flexibility and resiliency of the thin sheet metal in the seal areas aids in assuring maintenance of cell sealing.

Fig. 2 is a perspective exploded view of a fuel cell unit of a molten carbonate fuel cell stack according to one embodiment of this invention with separator plates 40, cathode 27, cathode current collector 28, electrolytes 20, anode 26 and anode current collector 29. Both separator plates 40 and electrolytes 20 extend to the edge of the cell and form wet seals on both faces of separator plates 40 around its entire periphery in peripheral wet seal areas 43. Peripheral wet seal areas 43 are extended both upwardly and downwardly from the general plane of separator plate 40 to provide contact with the periphery of electrolytes 20 on both faces of separator plate 40. Separator plates 40 and electrolyte tiles 20 are both penetrated by corresponding fuel manifold holes 24, one for supply and one for removal, and oxidant manifold holes 25, one for supply and one for removal. In the embodiment shown in Fig. 2, both separator plates 40 and electrolyte tiles 20 are penetrated only in their corner areas by manifold holes to provide the longest possible spacing between the manifold holes. As shown in Fig. 2, it may be preferred to have a manifold hole in each corner of separator plates 40 and electrolyte tiles 20.

While the manifold holes shown in Fig. 2 are a preferred triangular shape providing easily formed straight thin sheet manifold wet seal areas, the manifold holes may be round, rectangular or any other desired shape. The manifold holes shown in Fig. 2 are single openings, but partitions may be used in the single openings as desired to direct gas flow across the cell reactant chambers. Fuel manifold wet seal areas 45 and oxidant manifold wet seal areas 46 are extended both upwardly and downwardly from the general plane of separator plate 40 to provide contact with the electrolyte 20 on both faces of separator plate 40 to form wet seals with the adjacent electrolyte 20 defining the gas conduits. The surface of anode 26 is about flush with the level of peripheral wet seal 43 and oxidant manifold wet seal 46 to provide wet seal contact between separator plate 40 and electrolyte 20 in those areas. On the opposite face of separator plate 40, the surface of cathode 27 is about flush the level of peripheral wet seal 43 to provide fuel manifold wet seal 45 contact between separator plate 40 and electrolyte 20 in those areas.

As best seen in Fig. 2, oxidant manifold holes 25 are sealed by oxidant manifold wet seals 46 providing oxidant flow only to and from the cathode chamber (adjacent the upper face of the separator plate as shown) by oxidant supply openings 48 and oxidant exhaust openings 48' and preventing gas flow to or from the anode chamber while fuel manifold holes 24 are sealed by fuel manifold wet seals 45 providing fuel flow only to and from the anode chamber (adjacent the lower face of the separator plate as shown) by fuel supply openings 47 and fuel exhaust openings 47' and preventing gas flow to or from the cathode chamber. While the manifold wet seals are shown as straight pressed sheet metal structures, they can be any desired shape or structure to prevent gas flow. The manifold wet seals form a double wet seal between fuel manifold hole 24 and oxidant manifold hole 25.

Separator plates 40 may be comprised of suitable materials providing desired physical strength and gas separation. In many cell stacks it is preferred to use bimetallic separator plates in which stainless steel may be used on the cathode face and nickel or copper on the anode face to avoid ferrous metal corrosion. Separator plates may also be fabricated from ferrous alloys, such as type 300 series stainless steel alloys. The separator plates provide the dual function of providing a gas chamber non-reactive separator as well as providing structural strength to the fuel cell as an internal load bearing member. While it is preferred to use separator plates having a corrugated cross-sectional shape to provide both strength and better gas circulation adjacent the electrodes, the principles of this invention are also applicable to flat separator plates structured to provide peripheral wet seal areas and to provide wet seals around internal manifold

holes while allowing gas to pass to and from the internal manifolds as required for fuel cell operation. The fuel cell stack internal separator plates are desirably very thin sheets, in the order of about 0.010 inch.

Thin stamped stainless steel plates have been used in heat exchange technology as exemplified by the publications "Modern Designs For Effective Heat Transfer," American Heat Reclaiming Corp., 1270 Avenue of the Americas, New York, New York 10020 and "Superchanger Plate and Frame Heat Exchanger," Tranter, Inc. Wichita Falls, Texas 76307. These heat exchangers use a series of gasketed embossed or pressed metal plates bolted together between end frames to provide channels for passage of hot medium on one side of the plate and passage of cold medium on the other side of the plate. However, fuel cell stack separator plates present very different problems of sealing and corrosion under molten alkali metal carbonates fuel cell operating conditions and different manifold configuration, sealing, and fluid communication means since two fluids must pass in separated relation between adjacent separator plates. In heat exchange, only one fluid passes between adjacent heat exchange plates. However, the technology of fluid flow over the electrodes of the fuel cell stack of this invention may advantageously utilize design techniques and patterns of plate heat exchangers, such as herringbone, washboard, straight corrugations and mixed corrugations.

Fig. 3 shows in more detail a peripheral wet seal area in accordance with one embodiment of this invention wherein thin sheet separator plate 40 is corrugated with the peaks on one face of the corrugations adjacent cathode 27 support plate 28 with perforations 29 and formed to have flat thin sheet separator plate seal area 44 which lies adjacent electrolyte 20 on the cathode face of the cell. Separator plate wet seal strip 41 formed from thin metal strip material is welded by welds 42, or otherwise attached, to the anode face of separator plate 40 to provide flat separator plate wet seal strip wet seal area 43 which lies adjacent electrolyte 20 on the anode side of the cell. It is readily apparent that the position of the separator plate and the wet seal strip may be reversed and that spacing of separator wet seal strip wet seal area 43 and separator wet seal area 44 may be formed to fit the spacing requirements of individual cells.

Fig. 4 shows in cross-sectional view through a conduit between fuel manifold 24 and the anode chamber the manner in which separator plate fuel manifold wet seal area 45 between the lower face of separator plate 40 and electrolyte 20 prevents fuel flow to the cathode chamber and provides fuel flow to the anode chamber between anode 26 and the upper face of separator plate 40. Likewise, Fig. 5 shows in cross-sectional view through a conduit between oxidant manifold 25 and the cathode chamber the manner in which separator plate oxidant manifold wet seal

area 44 between the upper face of separator plate 40 and electrolyte 20 prevents oxidant flow to the anode chamber while providing oxidant flow to the cathode chamber between cathode 27 and the lower face of separator plate 40. The fuel and oxidant passages may be formed by corrugations in separator plate 40, by holes through a strip fastened to separator plate 40, or by any other means suitable to distribute the gases as desired.

Another embodiment of a separator plate according to this invention is shown in Figs. 6-10. In this embodiment, fuel supply and oxidant supply manifolds are arranged alternately along opposite ends of the thin sheet separator plate and spent fuel and spent oxidant manifolds are arranged alternately across a central region of the thin sheet separator plates to provide a split gas flow and greater mechanical stability of thin separator plates of large surface area. The thin metal separator plates are constructed in the same fashion as described above with pressed corrugations in the active areas to support the electrodes and to provide appropriate anode and cathode gas chamber volume and with pressed areas extending outwardly from the plane of the thin separator plate to form thin plate wet seal areas on one face of the plate and an attached outwardly extending thin formed sheet metal strip to form wet seal areas on the other face of the separator plate. Fig. 6 shows the top or front face of a separator plate while Fig. 7 shows the reverse face of the same separator plate. The electrochemically active areas of separator plate 140 are corrugated, as best seen in Fig. 9, with pressed periphery wet seal area 123 extending beyond the corrugations for contact with the electrolyte of one cell at periphery wet seal area 123, and pressed thin metal wet seal strip 141 attached to the periphery of the opposite face of separator plate 140 and extending beyond the corrugations to contact the electrolyte of the adjacent cell at periphery wet seal area 123. Oxidant manifold holes 125 are arranged alternately to fuel manifold holes 124 across opposite end regions and oxidant manifold holes 125A arranged alternately to fuel manifold holes 124A in the central region of separator plate 140. The series of oxidant manifold holes and fuel manifold holes, as shown in these figures, provide fuel and oxidant supply at opposite ends of separator plate 140 and fuel an oxidant removal in the central portion of separator plate 140. As best seen in Fig. 8, oxidant as supplied through manifold holes 125 and pass through oxidant supply openings 148 to the active surface of separator plate 140, as shown by the arrows in Fig. 6. The oxidant passes through the channels of corrugated separator plate 140 forming the cathode gas compartment to oxidant outlet openings 158, as shown by the arrows in Fig. 6, feeding oxidant manifold holes 125A. In a similar fashion, fuel is supplied through fuel manifold holes 124 to fuel supply openings 147, passes

through channels of corrugated separator plate 140 forming the anode gas compartment to fuel outlet openings 157, as shown by the arrows in Fig. 7, feeding fuel manifold holes 124A.

5 The co-linear flow of fuel and oxidant gases on opposite faces of the separator plate is shown in Figs. 6 and 7 with the supply manifolds at opposite ends of the separator plate and the outlet manifolds in a central region of the separator plate. Using the same separator plate reversed co-linear flow of fuel and oxidant gases on opposite faces of the plate may be obtained by using the central fuel and oxidant manifolds for supply and the end fuel and oxidant manifolds for outlets. Using the same separator plates counterflow of fuel and oxidant gases on opposite faces of the separator plate may be obtained by supplying one of the fuel or oxidant through the central fuel or oxidant manifolds while withdrawing the gas through corresponding outlet manifolds at both ends and introducing the other gas through the end manifolds and withdrawing through the central manifolds. It is thus seen that various desired gas flow patterns may be obtained on opposite faces of the separator plate through use of the identical separator plate and only altering the supply to the manifold or manifolds exterior to the cell.

20 We have found that by using thin metal sheet material in all wet seal areas of the separator plate, due to the limited flexibility and resiliency in the wet seal area upon assembly of the fuel cell stack minimization or prevention of gas leakage can be achieved across wet seals at least 1/4 inch wide separating adjacent fuel and oxidant manifolds. The thin sheet metal separator plates according to this invention show 25 good mechanical strength and provide ease of fabrication. The split flow design of separator plates as shown in Figs. 6-10, provides increased stiffness to the overall separator plate by the support of wet seal areas surrounding the manifold holes in the central portion of the plate. This design also allows fabrication of electrodes that are only a portion, in this case, half of the active current producing area of the fuel cells, facilitating handling of the electrodes and allowing continuous processing such as tape casting and sintering to be performed with smaller equipment. An 30 important feature of this invention is the flattened thin metal upstanding wet seal areas of the separator plate providing contact directly with the electrolyte of one cell on one face and the adjacent cell on the opposite face, the fuel and oxidant conduits passing 35 through only the separator plates and the electrolytes in the fuel cell stack.

40 Fig. 11 shows one embodiment of a fuel cell stack having unit cells as shown in Fig. 2 with a reforming chamber suitable to be interdispersed along the axis of the fuel cell stack. The reforming chamber is located 45 between anode separator/reformer plate 40" and cathode separator reformer plate 40'. Separator 50

plates 40, cathode 27, cathode current collector 28, electrolyte 20, anode 26 and anode current collector 29 are similar to those previously described with respect to Fig. 2 except for the provision of reactant gas manifold holes 50 and 52. Reactant gas manifold wet seal area 51 extends on both faces from the general plane of separator plate 40 to provide contact to form wet seals with the adjacent electrolyte 20 and define the reactant gas manifold, as described above with respect to the oxidant and fuel manifolds. Reactant gas manifold holes 50 are the same diameter while reactant gas manifold holes 52 have a larger diameter to enable the electrodes and current collectors to fit over extended reactant gas manifold wet seal area 51 to allow the flat surface of reactant gas manifold wet seal area 51 to contact electrolyte 20 for formation of a wet seal. The side walls of the extended reactant gas manifold wet seal areas are solid in separator plates 40 and therefore do not allow entry of reactant gas into the anode or cathode chambers. Anode separator/reformer plate 40" is different from separator plate 40 only in that oxidant supply openings 48 and oxidant exhaust openings 48' shown in separator plate 40 are not present and thus there is no communication between any of the oxidant manifolds and the reformer chamber adjacent the upper surface of anode separator/reformer plate 40". Reactant gas openings 53 in the sidewall of extended reactant gas manifold wet seal area 51 may be provided for communication between reactant gas manifold 50 and the reformer chamber. In a similar manner, the lower side of cathode separator/reformer plate 40' is modified by having solid sidewalls of extended spent fuel manifold wet seal area 45 to block communication between the reformer chamber and spent fuel manifold 24 by not providing fuel exhaust openings 47'. Reactant gas supply openings 53 may be provided for communication between reactant gas manifold 50 and the reformer chamber formed between anode separator/reformer plate 40" and cathode separator/reformer plate 40'. With these modifications to the standard separator plate 40, a reforming reaction chamber with fully internally manifolded reaction gas and steam supply may be interspersed at desired intervals in the fuel cell stack.

When the fuel cell stack is assembled, the peripheral wet seal area and all manifold wet seal areas of anode separator/reformer plate 40" come into contact with the corresponding wet seal areas extending from cathode separator/reformer plate 40' forming a tight metal/metal seal which, due to the limited flexibility and resiliency in the wet seal area, effectively seal the manifolds from the reformer chamber formed by the volume between plates 40" and 40'. If desired, the extended wet seal areas forming the reformer chamber may be further extended than on separator plates 40 to provide greater spacing between the separator/reformer plates and higher reformer chamber volume.

Another way to increase reformer chamber volume, if desired, is to provide sidewall extensions to each of the manifold wet seal areas and the peripheral wet seal area by welding such extensions to the separator/reformer plates providing any desired depth to the reformer chamber while providing closure of the reforming chamber and sealed passage of desired manifolds through the reformer chamber. When such additional depth is provided to the reformer chamber, metallic electrical conducting pillars may be provided at desired spaced locations between the separator/reformer plates to provide rigidity to the reformer chamber structure and to provide electrical continuity.

It is seen that the essential components for a fuel cell stack to have interspersed along its axis a plurality of reforming chambers is that each reforming chamber is formed by two spaced separator/reformer plates, one having an outer configuration of the face of the separator plate facing the anode chambers and the second having an outer configuration of the other face of the separator plate the cathode chambers, the two separator/reformer plates sealingly joined in their edge area to enclose a reformer chamber, conduits through the extended manifold wet seal structure providing reactant gas and steam communication from a reactant gas supply manifold to the reformer chamber, and conduits through the extended manifold wet seal structure providing enriched hydrogen product gas communication to a fuel gas supply manifold, thereby providing fully internal manifolding of reactant gas and steam to and product gas from each reformer chamber in the fuel cell stack.

Conventional reforming catalysts, such as supported nickel, may be used in the reformer chamber for conduct of the well known steam reforming reaction for the production of hydrogen from hydrocarbonaceous materials, such as natural gas or fuel obtained by gasification of naturally occurring organic carbonaceous materials such as coal, shale, or by anaerobic digestion. Any source of methane containing gas may be used to supply reactant hydrocarbonaceous gas to the reforming chambers.

As shown by the arrows, hydrocarbonaceous reaction gas and steam may be supplied from reactant gas manifold 50 to the reformer chamber, passed over reforming catalyst in the reforming chamber enhancing the hydrogen content of the product gas which is passed directly into fuel supply manifold 24.

In this manner, the hydrogen content of fuel passing through the fuel supply manifold may be enhanced along the axis of the cell.

The ability of fully internal manifolding a fuel cell stack as afforded by this invention allows interspersing of reforming chambers within the fuel cell stack which utilizes electrochemically generated heat to drive the reforming reaction while desirably cooling the electrochemical unit fuel cells and while providing

enhanced hydrogen fuel to the fuel supply manifold along the axis of the cell stack. Isolation of the reforming catalyst from the fuel cell electrolyte avoids poisoning of conventional reforming catalysts by molten carbonate electrolytes. The fully internally manifolded fuel cell stack with reforming of hydrocarbonaceous materials within the fuel cell stack provides improved overall fuel to electricity cell efficiency. According to this invention, natural gas may be used as a reactant to provide enhanced hydrogen fuel for the electrochemical reaction by placement of a reforming chamber, as described above, between about every five to about ten electrochemical unit cells.

While the use of reforming chambers has been described above with respect to the separator plate configuration shown in Fig. 2, it is clear that such reforming chamber configurations may be adopted to a wide variety of separator plate configurations, such as those shown in Figs. 6-10, by designation of certain of the internal manifolds as reactant gas manifolds with communication to a reforming chamber in a similar fashion to that described above. The essential element in provision of internal reforming chambers according to this invention is the fully internal manifolded fuel cell stack providing fuel supply and withdrawal, oxidant supply and withdrawal, and reactant gas supply through internal manifolds which may be arranged as desired and connected only through the end plates to external supply and exhaust conduits.

By use of separator plate/electrolyte wet seals, communication between the fuel manifold and only the anode face of the separator plate and between the oxidant manifold and only the opposite cathode face of the separator plate, can be achieved without porous gaskets as are essential when external manifolding is used. Additionally, reforming chambers may be interspersed along the axis of the fuel cell stack as described above. Further, each gas manifold seal area may be aluminized to reduce corrosive and other wicking processes.

Using the fully internal manifolding of this invention, the inter-cell changes of distance resulting from melting of the carbonates tape occurs at the factory assembly site and once such melting occurs there are no further changes in inter-cell distances. The height of the cell stack shipped from the factory will be the same as that during operation in a pressure vessel at the use site. The height of the reforming chambers will not change during start-up or fuel cell operation. Thus, the only follow up required during fuel cell stack operation is that required to maintain the cell holding force on the active and seal areas.

While in the foregoing specification this invention has been described in relation to certain preferred embodiments thereof, and many details have been set forth for purposes of illustration, it will be apparent to those skilled in the art that the invention is suscept-

ible to additional embodiments within the scope of the appended claims.

5 Claims

1. A generally rectangular fuel cell stack comprising a plurality of fuel cell units, each said fuel cell unit comprising an anode and a cathode, an electrolyte in contact with one face of said anode and an electrolyte in contact with an opposite facing face of said cathode, and a separator plate separating said cell unit between said anode and cathode forming an anode chamber between one face of said separator plate and said anode and a cathode chamber between the opposite face of said separator plate and said cathode, said anode chamber in gas communication with fuel gas supply and outlet and said cathode chamber in gas communication with oxidant gas supply and outlet the improvement comprising; said electrolytes and said separator plates extending to the edge of said fuel cell stack, said separator plates having a flattened peripheral wet seal structure extending to contact said electrolytes on each face of said separator plates completely around their periphery forming a separator plate/electrolyte wet seal under cell operating conditions, said electrolytes and said separator plates each having a plurality of aligned perforations said perforations in said separator plates being surrounded by a flattened manifold wet seal structure extending to contact said electrolyte on each face of said separator plate forming a separator plate/electrolyte wet seal under cell operating conditions to form a plurality of gas manifolds extending through said cell stack, conduits through said extended manifold wet seal structure providing fuel gas communication between one set of said manifolds and said anode chambers on one face of said separator plates and conduits through said extended manifold wet seal structure providing oxidant gas communication between a second set of said manifolds and said cathode chambers on the other face of said separator plates, thereby providing fully internal manifolding of fuel and oxidant gases to and from each said unit fuel cell in said fuel cell stack, said fuel cell stack having interspersed along its axis a plurality of reforming chambers each formed by two separator/reformer plates, one having the configuration of said one face of said separator plate facing one of said anode chambers and the second having the configuration of said other face of said separator plate facing one of said cathode chambers, said two separator/reformer plates sealingly joined in their edge area to enclose a reformer chamber, conduits through said

extended manifold wet seal structure providing reaction gas and steam communication from a third set of said manifolds to said reformer chamber, and conduits through said extended manifold wet seal structure providing enriched hydrogen product gas communication to a fuel gas supply manifold, thereby providing fully internal manifolding of reactant gas and steam to and product gas from each said reformer unit in said fuel cell stack.

2. In a fuel cell stack according to Claim 1 wherein end plates are configured the same as said separator plates on their inner faces and form half cells on each end of said fuel cell stack.

3. In a fuel cell stack according to Claim 1 wherein said separator and separator/reformer plates are pressed metal plates.

4. In a fuel cell stack according to Claim 1 wherein said flattened peripheral wet seal structure on one face of said plates comprises a pressed shaping of said plates to form said extended peripheral wet seal on said one face of said plates and on the other face of said plates comprises a pressed sheet metal shape forming said extended peripheral wet seal fastened to said other face of said plates.

5. In a fuel cell stack according to Claim 1 wherein said conduits through said extended manifold wet seal structure are formed by corrugated metal.

6. In a fuel cell stack according to Claim 1 wherein said conduits through said extended manifold wet seal structure are holes through sheet metal structures.

7. In a fuel cell stack according to Claim 1 wherein said flattened peripheral wet seal or extended manifold wet seal structure on one face of said separator plates and separator/reformer plates comprises a pressed shaping of said separator plates and separator/reformer plates to form said extended peripheral wet seal on said one face of said separator plates and separator/reformer plates and on the other face of said separator plates and separator/reformer plates comprises a pressed sheet metal shape forming said extended peripheral wet seal fastened to said other face of said separator plates and separator/reformer plates.

8. In a fuel cell stack according to Claim 1 wherein said reformer chambers are interspersed between groups of about five to about ten adjacent

5 fuel cell units.

9. In a fuel cell stack according to Claim 1 wherein said electrolyte comprises alkali metal carbonates.

10. A generally rectangular fuel cell stack comprising a plurality of fuel cell units, each said fuel cell unit comprising an anode and a cathode, an electrolyte in contact with one face of said anode and an electrolyte in contact with an opposite facing face of said cathode, and a separator plate separating said cell unit between said anode and cathode forming an anode chamber between one face of said separator plate and said anode and a cathode chamber between the opposite face of said separator plate and said cathode, said anode chamber in gas communication with fuel gas supply and outlet and said cathode chamber in gas communication with oxidant gas supply and outlet, said gas supplies and outlets fully internally manifolded within said fuel cell stack, a plurality of reforming chambers interspersed along the fuel cell stack axis each said reforming chamber formed by two separator/reformer plates, one having the configuration of one face of said separator plate facing one of said anode chambers and the second having the configuration of said other face of said separator plate facing one of said cathode chambers, said two separator/reformer plates sealingly joined in their edge area to enclose a reformer chamber, conduits through an extended manifold wet seal structure providing reaction gas and steam communication from an internal reactant gas manifold to said reformer chamber, and conduits through an extended manifold wet seal structure providing enriched hydrogen product gas communication to an internal fuel gas supply manifold, thereby providing fully internal manifolding of reactant gas and steam to and product gas from each said reformer unit in said fuel cell stack.

11. A process for production of electricity in a generally rectangular fuel cell stack comprising a plurality of fuel cell units, each said fuel cell unit comprising an anode and a cathode, an electrolyte in contact with one face of said anode and an electrolyte in contact with an opposite facing face of said cathode, and a separator plate separating said cell unit between said anode and cathode forming an anode chamber between one face of said separator plate and said anode and a cathode chamber between the opposite face of said separator plate and said cathode, comprising: passing fuel and oxidant gases through fully internal manifold conduits to and from each fuel cell unit in said fuel cell stack, said internal mani-

fold conduits formed by said electrolytes and said separator plates each having a plurality of aligned perforations, each said perforation being surrounded by a flattened manifold wet seal structure extending to contact said electrolyte on each face of said separator plate forming a separator plate/electrolyte wet seal under cell operating conditions to form a plurality of gas manifolds extending through said cell stack, conduits through said extended manifold wet seal structure providing fuel gas communication between one set of said manifolds and said anode chambers on one face of said separator plates and conduits through said extended manifold wet seal structure providing oxidant gas communication between the other set of said manifolds and said cathode chambers on the other face of said separator plates, thereby providing fully internal manifolding of fuel and oxidant gases to and from each said unit fuel cell in said fuel cell stack; and passing hydrocarbonaceous reactant gas to said fuel cell stack having interspersed along the fuel cell stack axis a plurality of reforming chambers, each formed by two separator/reformer plates, one having the configuration of said one face of said separator plate facing one of said anode chambers and the second having the configuration of said other face of said separator plate facing one of said cathode chambers, said two separator/reformer plates sealingly joined in their edge area to enclose a reformer chamber, conduits through said extended manifold wet seal structure providing reaction gas and steam communication from a third set of said manifolds to said reformer chamber passing said reactant gas over a reforming catalyst forming enriched hydrogen product gas, and passing said enriched hydrogen product gas through conduits through said extended manifold wet seal structure providing enriched hydrogen product gas to a fuel gas supply manifold, thereby providing fully internal manifolding of reactant gas and steam to and product gas from each said reformer unit in said fuel cell stack.

Patentansprüche

1. Ein annähernd rechteckiger Brennstoffzellenstapel, der eine Vielzahl von einzelnen Brennstoffzellen enthält, von denen jede eine Anode und eine Kathode enthält sowie einen Elektrolyten umfasst, auf dessen einer Seite die Anode aufliegt und dessen anderer Seite die Kathode aufliegt, wobei eine Separatorplatte, welche zwischen der Anode und der Kathode liegt, eine Anodenkammer auf einer Seite der Separatorplatte bildet, während die andere Seite der Separatorplatte ei-
- 5 2. Brennstoffzellenstapel nach Patentanspruch 1, dadurch gekennzeichnet, dass dessen Endplatten dieselbe Form wie die im Innern des Stapsels angeordneten Separatorplatten aufweisen und halbe Zellen bilden.
- 10 3. Brennstoffzellenstapel nach Patentanspruch 1, dadurch gekennzeichnet, dass die Separator-

ne Kathodenkammer bildet, wobei die Anodenkammer mit einer Brennstoffgaszufuhr und einem Auslass in Verbindung steht, während die Kathodenkammer mit einer Sauerstoffgaszufuhr und einem Auslass in Verbindung steht, dadurch gekennzeichnet, dass sich sowohl der Elektrolyt wie die Separatorplatten bis an den äusseren Rand des Brennstoffzellenstapels erstrecken, wobei die Separatorplatten peripher feuchte Dichtungsgebilde aufweisen, die unter Betriebsbedingungen flächenmässige feuchte Dichtungen zwischen dem Elektrolyt und der Separatorplatte bilden, wobei sich die Dichtungen entlang dem ganzen Umfang der Zelle erstrecken und wobei die Elektrolyte und die Separatorplatten je eine Anzahl von aufeinander ausgerichteten Durchlassöffnungen aufweisen, wobei die Durchlassöffnungen in den Separatorplatten von flächenmässigen feuchten Dichtungsbereichen umgeben sind, die je auf einer Seite der Separatorplatte am Elektrolyten anliegen, so dass sie im Betrieb eine Dichtung zwischen Separatorplatte und Elektrolyt und im Innern des Zellenstapels hindurchführenden Gaskanälen bilden, welche jeder Zelle des Brennstoffzellenstapels die Zufuhr von Brennstoffgas und Sauerstoffgas ermöglichen, wobei im Brennstoffzellenstapel entlang seiner Achse eine Anzahl von Umwandlungskammern zwischengeschaltet sind, die je aus zwei Separator/Umwandlungsplatten gebildet sind, von denen eine Platte die Form einer Seite der Separatorplatte aufweist, welche die Anodenkammer begrenzt, während die andere Platte die Form einer Seite der Separatorplatte aufweist, welche die Kathodenkammer begrenzt, wobei diese beiden Separator/Umwandlungsplatten am Umfang miteinander verbunden sind und die Umwandlungskammern bilden, wobei durch die erweiterten Kanäle mit feuchten Dichtungen geführte Leitungen aus einem dritten Satz von Kanälen Reaktionsgas und Dampf der Umwandlungskammer zuführen, leiten durch die erweiterten Kanäle hindurchgeführte Leitungen mit Wasserstoff angereichertes Brennstoffgas in den Brennstoff-Zuführkanal, derart, dass Reaktionsgas und Dampf durch gänzlich im Innern geführte Kanäle den im Brennstoffzellenstapel angeordneten Umwandlungseinheiten zugeführt und Produktionsgas abgeleitet wird.

platten aus Metall gepresste Platten sind.

4. Brennstoffzellenstapel nach Patentanspruch 1, dadurch gekennzeichnet, dass die Gebilde, welche die feuchten Dichtungen am Umfang bilden, an einer Seite der Separatorplatte aus einem mit der Separatorplatte verbundenen gepressten Metallstreifen bestehen, welcher die erweiterte feuchte Dichtung auf der anderen Seite der Separatorplatte bildet.
5. Brennstoffzellenstapel nach Patentanspruch 1, dadurch gekennzeichnet, dass die Leitungskanäle durch die erweiterten feuchten Dichtungsbereiche aus gewelltem Metallblech geformt sind.
10. Brennstoffzellenstapel nach Patentanspruch 1, dadurch gekennzeichnet, dass die Leitungskanäle durch die erweiterten feuchten Dichtungsbereiche aus Oeffnungen in den Gebilden aus Blech bestehen.
15. Brennstoffzellenstapel nach Patentanspruch 1, dadurch gekennzeichnet, dass die Gebilde, welche die feuchten, flachen Dichtungen am Umfang oder die feuchte Dichtung der erweiterten Kanäle an einer Seite der Separatorplatten oder an einer Seite der Separator/Umwandlerplatten bilden aus einer aus dem Plattenmaterial gepressten Form bestehen, während an der anderen Seite der Separatorplatten oder an der anderen Seite der Separator/Umwandlerplatte ein aus Metall gepresstes Gebilde angebracht ist, welches mit der genannten anderen Seite der Separatorplatten oder Separator/Umwandlerplatte verbunden ist.
20. Brennstoffzellenstapel nach Patentanspruch 1, dadurch gekennzeichnet, dass jeweils zwischen Gruppen von fünf bis zehn Brennstoffzellen eine Umwandlungskammer angebracht ist.
25. Brennstoffzellenstapel nach Patentanspruch 1, dadurch gekennzeichnet, dass das Elektrolyt Alkalimetallkarbonate enthält.
30. Ein annähernd rechteckiger Brennstoffzellenstapel, der eine Vielzahl von einzelnen Brennstoffzellen enthält, von denen jede eine Anode und eine Kathode enthält sowie einen Elektrolyten umfasst, auf dessen einer Seite die Anode aufliegt und auf seiner anderen Seite die Kathode aufliegt, wobei eine Separatorplatte, welche zwischen der Anode und der Kathode liegt, eine Anodenkammer auf einer Seite der Separatorplatte bildet, während die andere Seite der Separatorplatte eine Kathodenkammer bildet, wobei die Anodenkammer mit einer Brennstoffgaszufuhr

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und einem Auslass in Verbindung steht, während die Kathodenkammer mit einer Sauerstoffgaszufuhr und einem Auslass in Verbindung steht, wobei die Gaszufuhr- und Ableitungskanäle vollständig im Innern des Zellenstapels verlaufen und eine Vielzahl von Umwandlungskammern entlang der Achse des Stapes zwischengefügt ist, die je aus zwei Separator/Umwandlungsplatten bestehen, von denen eine dieser Platten zur Anodenkammer, die andere dieser Platten zur Kathodenkammer hin gerichtet ist, wobei die zwei Separator/Umwandlungsplatten an ihrem Umfang dicht miteinander verbunden sind und die Umwandlungskammer umschließen, wobei Leitungen, welche durch die erweiterte Durchlassöffnung mit feuchter Dichtung der Kanäle hindurchführen, der Umwandlungskammer Reaktionsgas und Dampf aus einem im Innern geführten Reaktionsgaskanal zuführen, während Leitungen, welche durch die erweiterte Durchlassöffnung mit feuchter Dichtung mit Wasserstoff angereichertes Reaktionsgas in den im Innern geführten Brennstoffgaskanal leiten und auf diese Weise durch vollständig im Innern geführte Kanäle der Umwandlungskammer Reaktionsgas und Dampf zuführen und den Zellen des Brennstoffzellenstapels Produktionsgas zuführen.

11. Verfahren zum Produzieren von Elektrizität in einem annähernd rechteckigen Zellenstapel, der eine Vielzahl von Brennstoffzellen enthält, wobei jede Brennstoffzelle eine Anode und eine Kathode enthält sowie einen Elektrolyten, auf dessen einer Seite die Anode und einen Elektrolyten, auf dessen anderer Seite die Kathode aufliegt, wobei eine Separatorplatte zwischen der Anode und der Kathode liegt eine Anodenkammer zwischen der Anode und einer Seite der Separatorplatte bildet, während zwischen der anderen Seite der Separatorplatte und der Kathode eine Kathodenkammer gebildet wird, dadurch gekennzeichnet, dass sämtlichen Zellen des Brennstoffzellenstapels gasförmiger Brennstoff und Sauerstoff durch die im Innern geführten Kanäle zugeleitet und abgeführt wird, wobei diese Kanäle und Separatorplatten aus dem Elektrolyten und den Separatorplatten gebildet sind, indem diese mit einer Vielzahl von aufeinander ausgerichteten Durchbrüchen versehen sind, die durch ein Gebilde mit flachem Rand umgeben sind, welcher bis zum Elektrolyt reicht und beim Betrieb eine feuchte Dichtung zwischen der Separatorplatte und dem Elektrolyt bildet und so eine Vielzahl von durch den Zellenstapel hindurchführenden, feucht abgedichteten Kanäle bildet, von denen ein Satz der Verbindung mit den Anodenkammern an einer Seite der Separatorplatte dient und Brennstoffgas zuführt, während ein anderer Satz von Kanälen die Ver-

bindung mit den Kathodenkammern an der anderen Seite der Separatorplatte herstellt und Sauerstoffgas zuführt, so dass die im Innern geführten Kanäle jeder Zelle des Brennstoffzellenstapels Brennstoff und Sauerstoff zuführen und abführen, wobei kohlenwasserstoffhaltiges Reaktionsgas zu einer Vielzahl von im Brennstoffzellenstapel in dessen Längsrichtung eingefügten Umwandlungskammern geführt wird, die je aus zwei Separator/Umwandlungsplatten bestehen, von denen eine die Form einer Separatorplatte aufweist, welche eine Anodenkammer begrenzt und die andere die Form einer Separatorplatte aufweist, welche eine Kathodenkammer begrenzt, wobei diese beiden Separator/Umwandlerplatten entlang ihrem Umfang gasdicht miteinander verbunden sind und so die Umwandlungskammer umschließen.

Durchlässe in einem dritten Satz von erweiterten Kanälen stellen eine Verbindung von Reaktionsgas zur Umwandlungskammer her, in welcher das Reaktionsgas über einen Umwandlungskatalysator strömt und ein mit Wasserstoff angereichertes Produktionsgas erzeugt wird, welches durch die erweiterten Kanäle mit feuchter Abdichtung in den Brennstoffgaszuführkanal gelangt, derart, dass die im Innern geführten Kanäle sämtlichen im Brennstoffzellenstapel angebrachten Umwandlungskammern Reaktionsgas und Dampf zuführen und Produktionsgas aus denselben abführen.

Revendications

1. Empilement de piles à combustible généralement rectangulaire, comprenant une pluralité d'unités de piles à combustible, chacune des dites unités de pile à combustible comprenant une anode et une cathode, un électrolyte en contact avec une face de ladite anode et un électrolyte en contact avec une face de ladite cathode orientée en regard, et une plaque séparatrice séparant ladite unité de pile entre ladite anode et ladite cathode, formant une chambre d'anode entre une face de ladite plaque séparatrice et ladite anode et une chambre de cathode entre la face opposée de ladite plaque séparatrice et ladite cathode, ladite chambre d'anode étant en communication pour les gaz avec une alimentation en gaz combustible et une sortie et ladite chambre de cathode étant en communication pour les gaz avec une alimentation en gaz oxydant et une sortie, comprenant le perfectionnement selon lequel: les dits électrolytes et les dites plaques séparatrices s'étendent jusqu'au bord du dit empilement de piles à combustible, les dites plaques séparatrices ayant une structure de scellement humide périphérique aplatie, s'éten-

dant de façon à être en contact avec les dits électrolytes sur chaque face des dites plaques séparatrices sur toute leur périphérie, de façon à former un scellement humide plaque séparatrice/électrolyte dans les conditions de fonctionnement des piles, les dits électrolytes et les dites plaques séparatrices, dont chacun a une pluralité de perforations alignées, les dites perforations dans les dites plaques séparatrices étant entourées par une structure de scellement humide à canaux aplatis, s'étendant de façon à être en contact avec le dit électrolyte sur chaque face de la dite plaque séparatrice formant une étanchéité humide plaque séparatrice/électrolyte dans les conditions de fonctionnement de la pile pour former une pluralité de tubulures de gaz s'étendant à travers ledit empilement de piles, des conduites à travers la dite structure de scellement humide à canaux étendus procurant une communication de gaz combustible entre un groupe des dites tubulures et les dites chambres d'anode sur une face des dites plaques séparatrices et des conduites à travers la dite structure de scellement humide à canaux étendus procurant une communication de gaz oxydant entre un second groupe des dites tubulures et les dites chambres de cathode sur l'autre face des dites plaques séparatrices, fournissant ainsi une distribution totalement interne des gaz combustible et oxydant vers et à partir de chaque unité de pile à combustible susdite dans ledit empilement de piles à combustible, ledit empilement de piles à combustible ayant, intercalées le long de son axe, une pluralité de chambres de reformage formées chacune par deux plaques séparatrices/reformeuses, l'une ayant la configuration de l'une des dites faces de la dite plaque séparatrice faisant face à l'une des dites chambres d'anode et la seconde ayant la configuration de la dite autre face de la dite plaque séparatrice faisant face à l'une des dites chambres de cathode, les dites deux plaques séparatrices/reformeuses formant un joint étanche dans leur zone de bord pour constituer une chambre de reformage, des tubulures dans la dite structure de scellement humide à canaux étendus procurant une communication du gaz de réaction et de la vapeur à partir d'un troisième ensemble de tubulures susdites vers la dite chambre de reformage, et des tubulures à travers la dite structure à scellement humide à canaux étendus procurant une communication de gaz hydrogène enrichi produit vers une tubulure d'alimentation en gaz combustible, fournissant ainsi une distribution totalement interne du gaz réactif et de la vapeur vers et du gaz produit à partir de chacune des dites unités de reformage dans le dit empilement de piles à combustible.

2. Empilement de piles à combustible selon la revendication 1, dans lequel les plaques d'extrémité sont configurées de la même manière que les dites plaques séparatrices sur leurs faces internes et forment des demi-piles sur chaque extrémité du dit empilement de piles à combustible.

3. Empilement de piles à combustible selon la revendication 1, dans lequel le dit séparateur et les dites plaques séparatrices/reformées sont des plaques en tôle emboutie.

4. Empilement de piles à combustible selon la revendication 1, dans lequel la dite structure de scellement humide périphérique aplatie sur une face des dites plaques comprend une mise en forme emboutie des dites plaques pour former le dit scellement humide périphérique étendu sur la première face des dites plaques et sur l'autre face des dites plaques comprend une forme de métal en feuille emboutie formant le dit scellement humide périphérique étendu, fixée à la dite autre face des dites plaques.

5. Empilement de piles à combustible selon la revendication 1, dans lequel les dites tubulures à travers la dite structure de scellement humide à canaux étendus sont formées par du métal onduillé.

6. Empilement de piles à combustible selon la revendication 1, dans lequel les dites tubulures à travers la dite structure de scellement humide à canaux étendus sont des trous au travers des structures métalliques en feuille.

7. Empilement de piles à combustible selon la revendication 1, dans lequel la dite structure de scellement humide périphérique aplatie ou structure de scellement humide à canaux étendus comprend sur une des faces des dites plaques séparatrices et plaques séparatrices/reformées un profilage embouti des dites plaques séparatrices et plaques séparatrices/reformées pour former le dit scellement humide périphérique étendu sur la dite première face des dites plaques séparatrices et plaques séparatrices/reformées et comprend sur l'autre face des dites plaques séparatrices et plaques séparatrices/reformées un profil de feuille métallique emboutie formant le dit scellement humide périphérique étendu, fixé à la dite autre face des dites plaques séparatrices et plaques séparatrices/reformées.

8. Empilement de piles à combustible selon la revendication 1, dans lequel les dites chambres reformées sont intercalées entre des groupes

5 d'environ cinq à environ dix unités de piles à combustible adjacentes.

9. Empilement de piles à combustible selon la revendication 1, dans lequel le dit électrolyte comprend des carbonates de métal alcalin.

10. Empilement de piles à combustible généralement rectangulaire, comprenant une pluralité d'unités de piles à combustible, chacune des dites unités de piles à combustible comprenant une anode et une cathode, un électrolyte en contact avec une face de ladite anode et un électrolyte en contact avec une face de ladite cathode orientée en regard, et une plaque séparatrice séparant ladite unité de pile entre ladite anode et ladite cathode, formant une chambre d'anode entre une face de ladite plaque séparatrice et ladite anode et une chambre de cathode entre la face opposée de ladite plaque séparatrice et ladite cathode, ladite chambre d'anode étant en communication pour les gaz avec une alimentation en gaz combustible et une sortie et ladite chambre de cathode étant en communication pour les gaz avec une alimentation en gaz oxydant et une sortie, les dites alimentations en gaz et sorties de gaz étant totalement canalisées intérieurement dans le dit empilement de piles à combustible, une pluralité de chambres de reformage intercalées le long de l'axe de l'empilement de cellules à combustible, chacune des dites chambres de reformage étant formée par deux plaques séparatrices/reformées l'une ayant la configuration d'une face de la dite plaque séparatrice faisant face à l'une des dites chambres d'anode et la seconde ayant la configuration de la dite autre face de la dite plaque séparatrice faisant face à l'une des dites chambres de cathode, les dites deux plaques séparatrices/reformées étant réunies de manière étanche dans la zone de leurs bords pour enclore une chambre reformée, des tubulures à travers une structure de scellement humide à canaux étendus procurant une communication de gaz réagissant et de vapeur entre des canaux internes pour gaz réagissant et la dite chambre reformée, et des tubulures traversant une structure de scellement humide à canaux étendus procurant une communication du gaz hydrogène enrichi produit avec des canaux internes d'alimentation en gaz combustible, ce qui procure une distribution totalement interne en gaz réagissant et en vapeur vers et en gaz produit à partir de chacune des dites unités reformées dans ledit empilement de piles à combustible.

11. Procédé pour la production d'électricité dans un empilement de piles à combustible généralement

rectangulaire comprenant une pluralité d'unités de piles à combustible, chacune des dites unités de piles à combustible comprenant une anode et une cathode, un électrolyte en contact avec une face de ladite anode et un électrolyte en contact avec une face de ladite cathode orientée en regard, et une plaque séparatrice séparant ladite unité de pile entre ladite anode et ladite cathode, formant une chambre d'anode entre une face de ladite plaque séparatrice et ladite anode et une chambre de cathode entre la face opposée de ladite plaque séparatrice et ladite cathode, comprenant le passage de gaz combustible et de gaz oxydant à travers des tubulures totalement internes vers et depuis chaque unité de pile à combustible dans le dit empilement de piles à combustible, les dites tubulures internes formées par les dits électrolytes et les dites plaques séparatrices ayant chacune une pluralité de perforations alignées, chacune des dites perforations étant entourée par une structure de scellement humide à canaux aplatis s'étendant pour entrer en contact avec le dit électrolyte sur chaque face de la dite plaque séparatrice formant un scellement humide plaque séparatrice/électrolyte dans les conditions de fonctionnement de la pile pour former une pluralité de canaux pour les gaz s'étendant à travers le dit empilement de piles, des tubulures traversant la dite structure de scellement humide à canaux étendus procurant une communication pour le gaz combustible entre un groupe des dits canaux et les dites chambres d'anode sur une face des dites plaques séparatrices et des tubulures traversant la dite structure de scellement humide à canaux étendus procurant une communication de gaz oxydant entre l'autre groupe des dits collecteurs et les dites chambres de cathode sur l'autre face des dites plaques séparatrices, procurant ainsi une canalisation totalement interne des gaz combustible et oxydant vers et depuis chacune des dites unités de piles à combustible dans le dit empilement de piles à combustible; et le passage de gaz réactif hydrocarboné dans le dit empilement de piles à combustible ayant, intercalé le long de l'axe de l'empilement de piles à combustible, une pluralité de chambres de reformage, chacune formée par deux plaques séparatrices/reformeuses, l'une ayant la configuration de la dite face de la dite plaque séparatrice faisant face à l'une des dites chambres d'anode et la seconde ayant la configuration de la dite autre face de la dite plaque séparatrice faisant face à l'une des dites chambres de cathode, les deux dites plaques séparatrices/reformeuses étant réunies de façon étanche à proximité de leurs bords pour contenir une chambre de reformage, des tubulures à travers la dite structure de scellement humide à canaux

5 étendus procurant une communication de gaz réactif et de vapeur depuis un troisième ensemble de canaux jusqu'à la dite chambre de reformage faisant passer le dit gaz réactif sur un catalyseur de reformage pour donner lieu à une production de gaz hydrogène enrichi, et le passage du dit gaz hydrogène enrichi produit dans des conduites traversant la dite structure de scellement humide à canaux étendus fournissant le gaz hydrogène enrichi produit à un collecteur d'alimentation en gaz combustible, procurant ainsi une distribution totalement interne de gaz réactif et de vapeur dans et de gaz produit à partir de la dite unité de reformage dans le dit empilement de piles à combustible.

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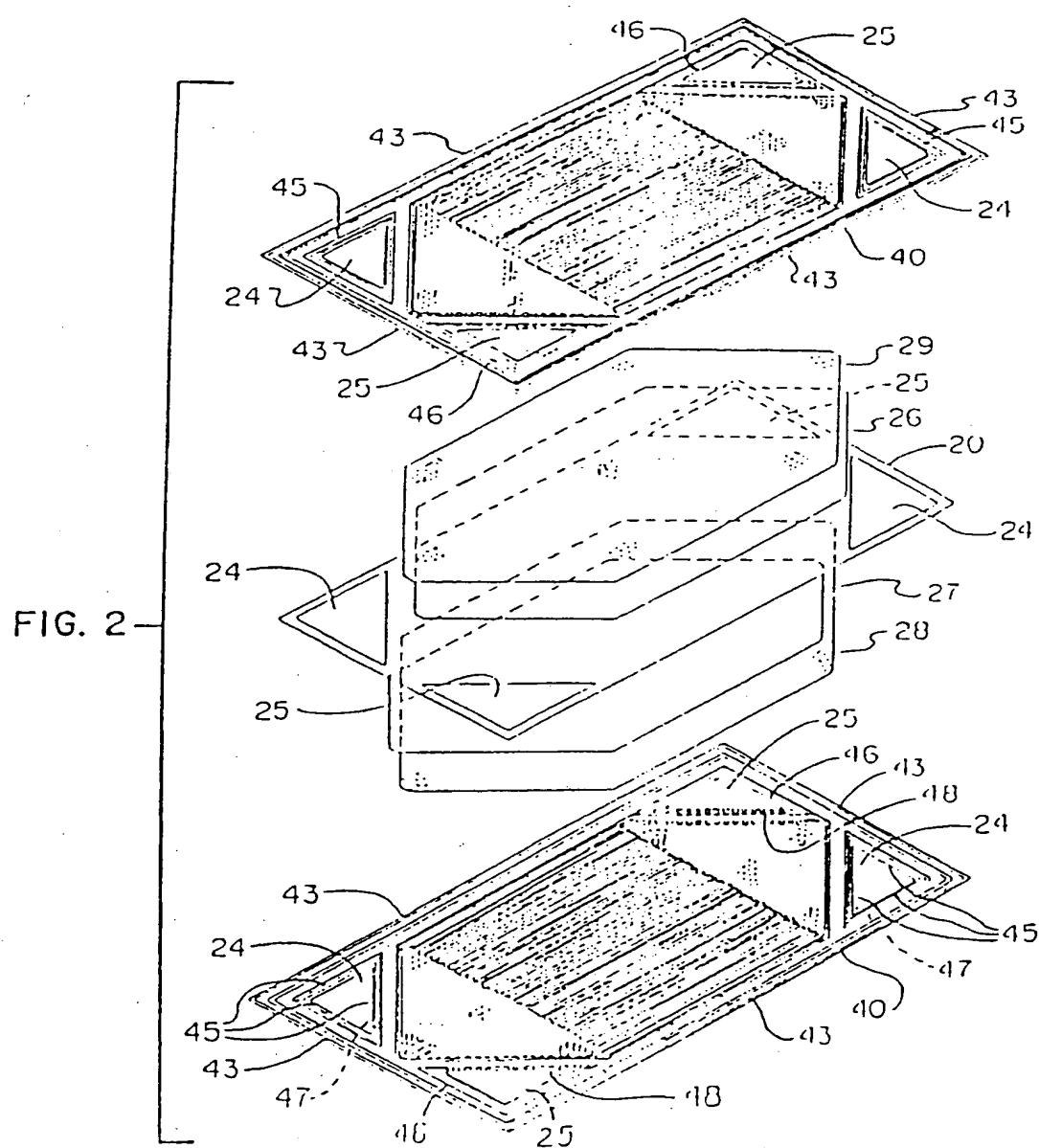
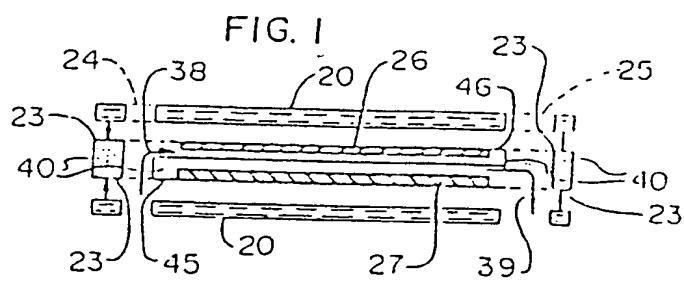
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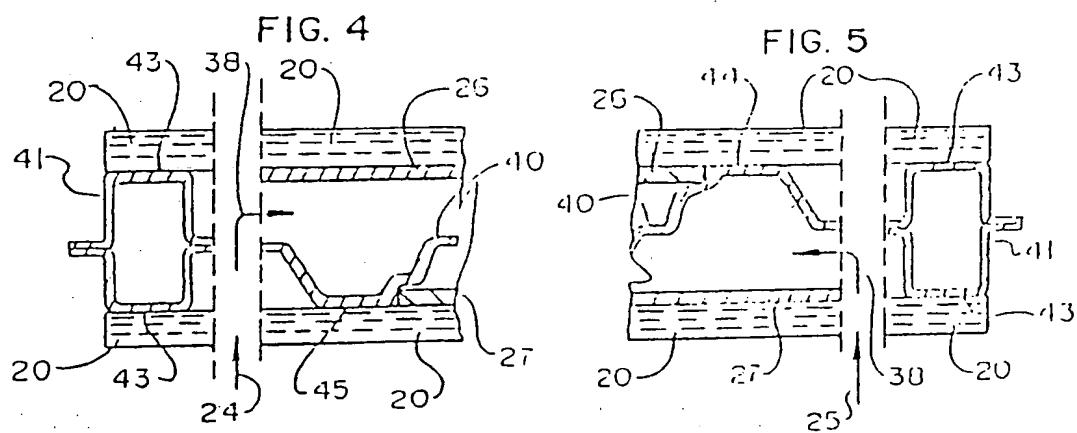
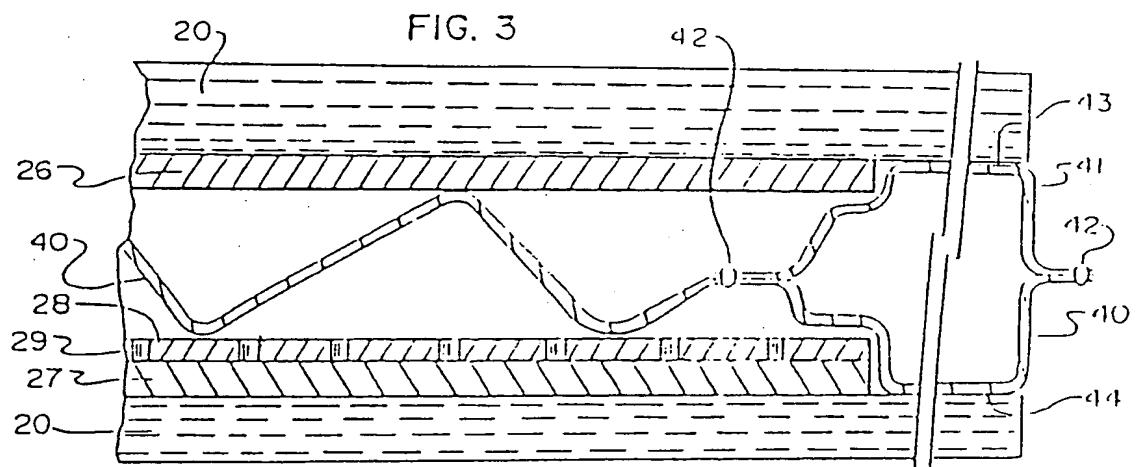


FIG: 7

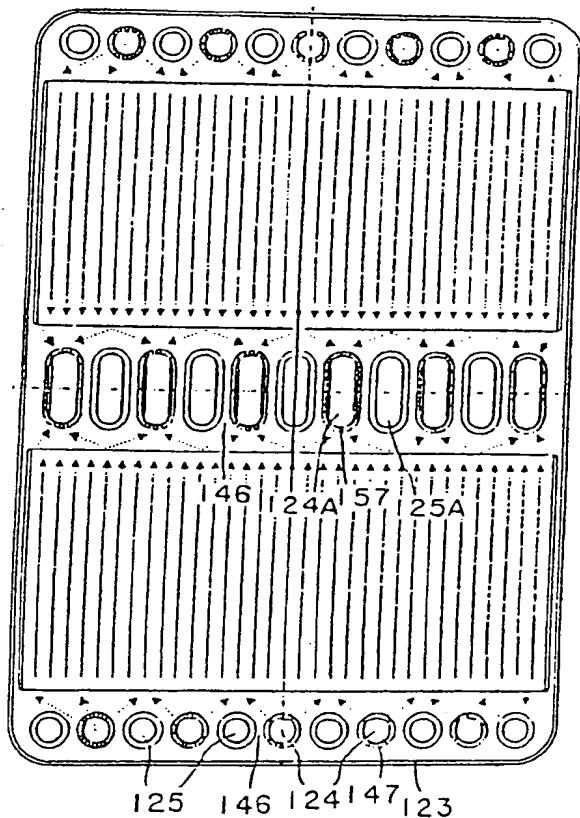


FIG. 6

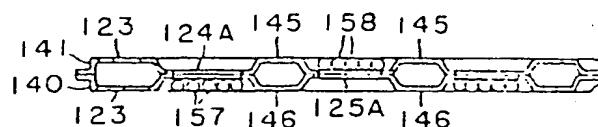
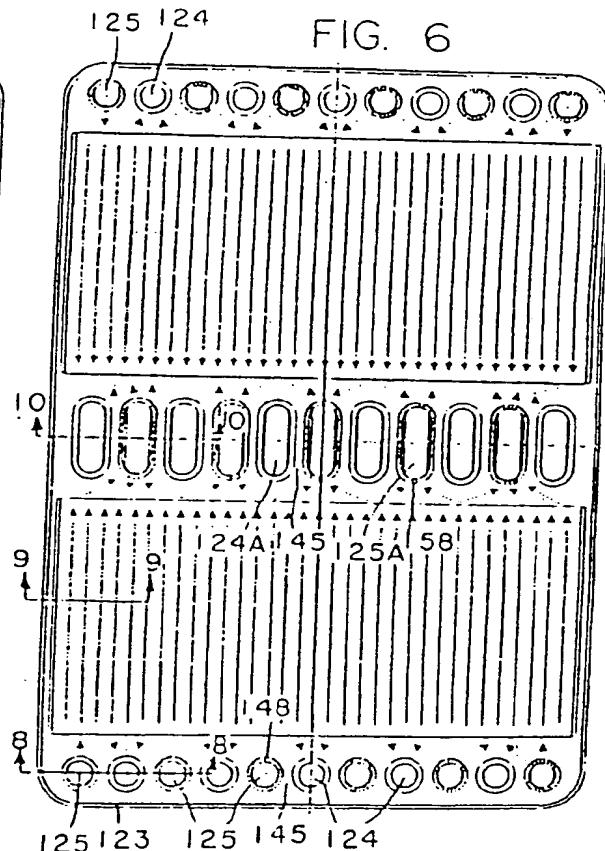


FIG. 10

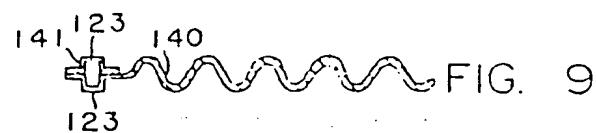


FIG. 9

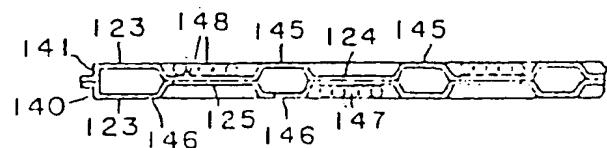
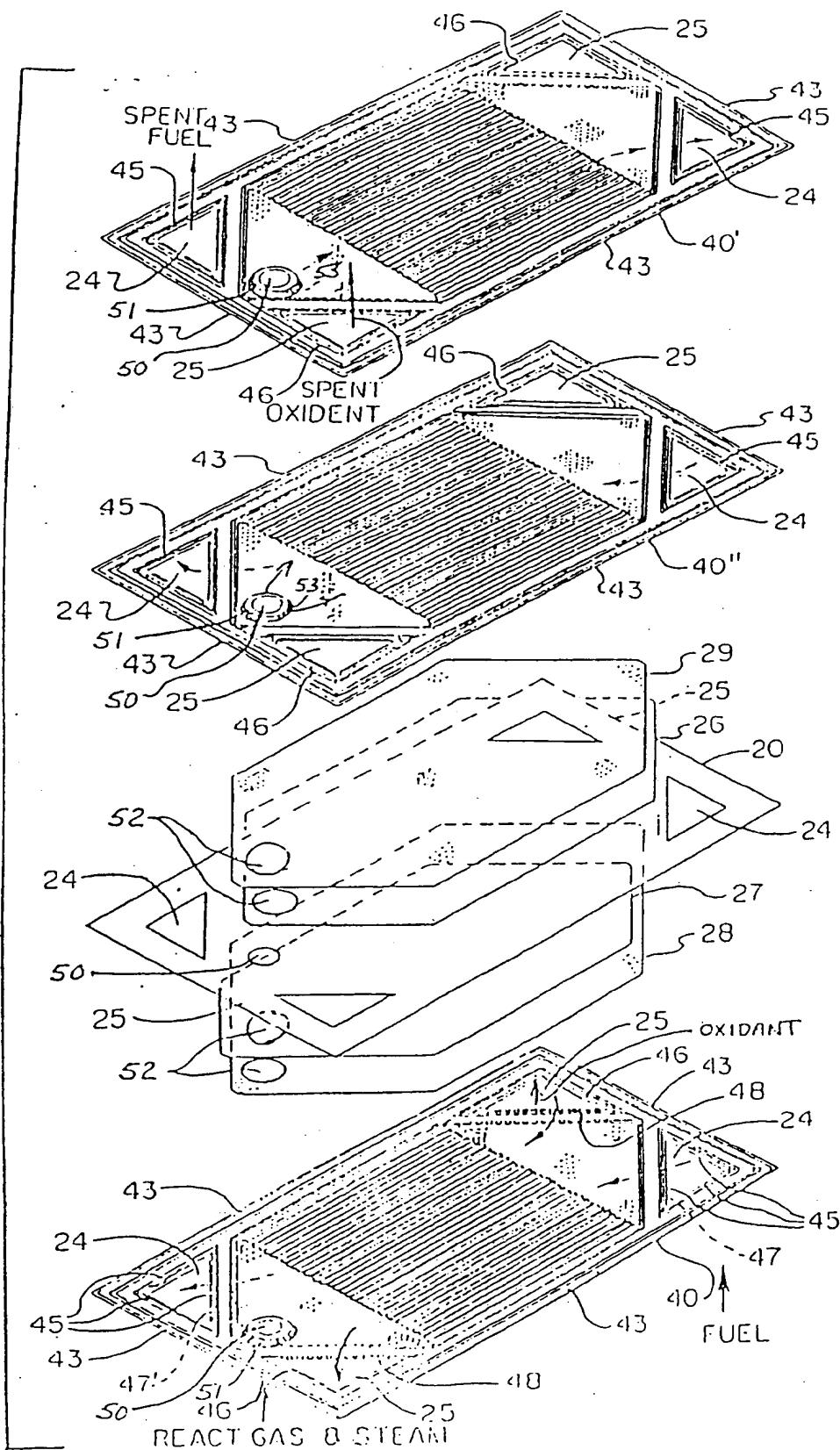


FIG. 8

FIG. II -



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